An Evaluation of Results for Samples Collected at LBNL in Accordance with the 2001 Vegetation Sampling Plan for Tritium

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ABSTRACT

This report evaluates data collected in accordance with the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001), compares the data with samples collected in the past, and evaluates whether there is a potential for adverse impact on the environment or human health from the presence of tritium in vegetation as a result of LBNL operations.

The distribution of environmental tritium in the tissue free water tritium (TFWT) and organically bound tritium (OBT) of tree wood, leaves, duff (material such as leaves, bark, and twigs that accumulates on the ground), and transpired water in the vicinity of Lawrence Berkeley National Laboratory (LBNL) has been evaluated. These measurements indicate that concentrations of both TFWT and OBT were at or below the minimum detectable activity in vegetation growing at distances greater than a few hundred meters from the National Tritium Labeling Facility (NTLF) hillside stack.

This document considers plausible scenarios that could result in human exposure. The maximum individual dose from tritium exposure calculated in this report, which occurs in a scenario involving processing of wood and leaves into mulch, is about 8×10^{-2} mrem. The maximum collective dose, which occurs in the scenario involving use of wood in domestic fireplaces, is about 2×10^{-3} person-rem. Based on these doses, the expected number of cancers from tritium released from LBNL in the exposed population is zero.

Conservative bounding calculations were completed to estimate the amount of tritium being released from the hillside grove through transpiration and the resulting dose and risk for a maximally exposed individual at the LHS (Lawrence Hall of Science). The estimated dose that an employee of the LHS for 30 years would receive from exposure to tritium being released from the hillside grove through transpiration would be less than 0.004 mrem. The lifetime risk from this dose would be less than 4×10^{-9}.

The detectable quantities of tritium in vegetation surrounding LBNL are far below levels at which there would be reason for concern for adverse impacts to the environment or human health. Exposure to tritium contained in vegetation surrounding LBNL should result in lifetime doses to maximally exposed individuals of less than 1 mrem, considering plausible routes and mechanisms of human exposure.

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1.0 BACKGROUND

In 1996, the City of Berkeley requested independent soil and vegetation sampling for HTO and organically bound tritium (OBT) at the site of the Lawrence Hall of Science (LHS). The U.S. Environmental Protection Agency (EPA) subsequently requested additional information on potential environmental hazards from Berkeley Lab.

Over the past four years, Berkeley Lab has taken hundreds of vegetation samples and has obtained a large set of data pertaining to the presence of tritium in vegetation near the National Tritium Labeling Facility (NTLF). In 2001, a written plan for sampling tritium in vegetation was developed (LBNL, 2001) to further characterize tritium concentrations in tree wood, leaves, duff, and transpired water resulting from releases from the Berkeley Lab hillside stack.

1.1 Vegetation Sampling Locations

The 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001) identifies general areas where vegetation samples are to be collected. Within these general areas, the plan specifies that samples of leaves, wood, duff, and transpired water are to be guided by the following considerations:

- All locations and distances were to be off-site, but as close as possible to the LBNL site boundaries, and where trees and vegetation can be found.

- Near the NTLF stack, predominant wind directions are toward the east-southeast and northwest. Since the population density in the south-southeast direction is extremely low, only one sample site is located in this direction. More samples (five) were collected to the northwest, north-northwest, and north of the Lawrence Hall of Science.

- Environmental tritium levels on-site are elevated near the NTLF hillside stack and decrease with distance from the stack. Based on the results of previous extensive vegetation tritium sampling, only trees within a radius of 200 meters of the NTLF stack have tritium concentration levels above levels of analytical detection. Half of the sampling locations are within 200 meters of the NTLF stack.

- The availability of vegetation (i.e., available mature trees at a location, available appropriate species, and accessible vegetation for sampling) must be suitable.

The locations at which vegetation samples were actually taken are listed in Table 1 and shown in Figure 1. Two sampling locations were selected to represent background measurements of tritium in vegetation, one approximately 20 km southeast (SEE9) of the NTLF stack and one approximately 1 km northeast (NEE10); these locations are not included in Figure 1. Five locations (NNW1, NNW2, NNW3, WNW4, and NNN5) were selected to provide samples in the vicinity of the Lawrence Hall of Science. One sampling location (EEE6) was selected at the east site boundary. Another location (SSE7) was selected in the south-southeast direction. Sample location WWW8 is at the site boundary directly west of the NTLF stack. Samples of wood, duff, and leaves were taken at all locations. Transpired water was measured at NNW1, NNW2, NNW3, and the two background locations (SEE9 and NEE10).

Actual sampling locations were at or near the general proposed locations (Table 1). Any differences between proposed and actual sample locations are due to physical limitations imposed by prevailing conditions in the field and were reviewed with EPA and DOE representatives. The actual sampling locations fully meet the requirements of the vegetation sampling plan and have been reviewed by EPA and reviewed and approved by DOE (Bandrowski, 2002 and Nolan, 2001).

1.2 Tissue Free Water Tritium vs. Organically Bound Tritium

Tritiated water vapor released to the environment readily mixes and exchanges with atmospheric water, such as precipitation, fog and vapor, and other sources of environmental water such as plant water, surface water, and soil water. Within vegetation, tritium exists as either tissue free-water tritium (TFWT) or organically bound tritium (OBT).

TFWT (or unbound tritium) is taken up rapidly by plants and is lost or turned over very rapidly through exchange with water vapor in the atmosphere. TFWT is tritium that can be removed from the plant by azeotropic distillation or freeze-drying.

Table 1. Description of sampling locations selected in accordance with the 2001 Vegetation Sampling Plan for Tritium.

	Proposed Sample Locations		**Actual Sample Locations**		
Location Number	**Direction from NTLF Stack**	**Distance from NTLF Stack (m)**	**Direction from NTLF Stack**	**Distance from NTLF Stack (m)[a]**	**Tree Diameter (cm)**
NNW1	NNW	20	NNW	27[b]	48-52[c]
NNW2	NNW	100	NNW	105 ± 12	40-47
NNW3	NNW	300	NNW	363 ± 7	55-68
WNW4	WNW	100	WSW	103 ± 10	39-44.5[c]
NNN5	N	50	N	45 ± 6	46-49[c]
EEE6	E	200	E	215 ± 8	49-51
SSE7	SSE	600	SSE	529 ± 8	53-70
WWW8	W	850	W	832 ± 34	46-49
SEE9	SE	20,000	SE	20,800 ± 12	36-40
NEE10	NE	1,000	NE	1080 ± 12	34-37

[a] Distances from NTLF stack are based on global positioning satellite (GPS) readings. The uncertainty represents the 95% uncertainty range associated with the use of a Garmin GPS unit.

[b] Actual measurement taken with a tape measure.

[c] Diameter of tree sampled for wood and duff; leaves were not accessible, so a nearby, smaller tree was sampled for leaves and transpired water

OBT is usually formed when tritium originating with tissue free water becomes incorporated into organic molecules. OBT is defined as the tritium fraction that remains after free water tritium is removed and consists primarily of tritium that is chemically bonded to organic compounds such as cellulose and sugars.

OBT is retained in plants for a much longer time than TFWT. TFWT usually represents the recent exposure of plants to HTO while OBT may represent time integration of past exposures to HTO (Murphy, 1993).

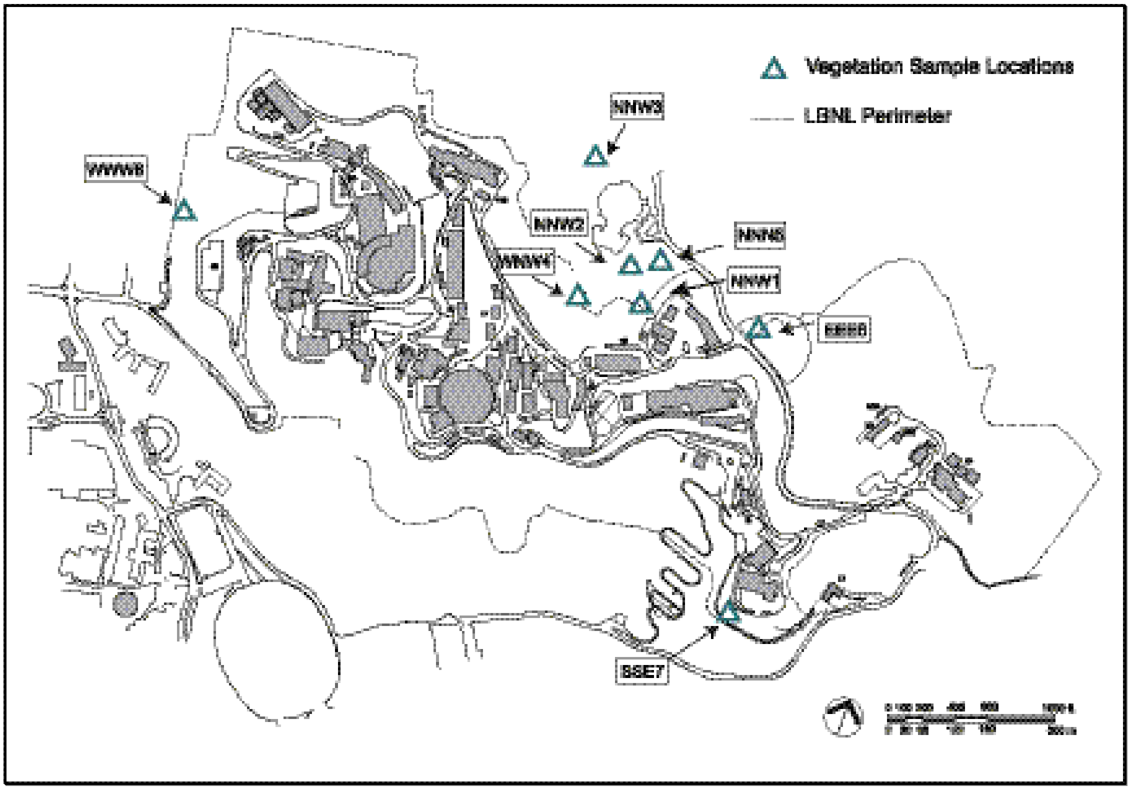

Figure 1. Actual Vegetation Sampling Locations for Tritium

1.3 Wet Season vs. Dry Season Sampling

Samples were collected in 2001 at the above mentioned locations during two separate time periods: September and November - January. These time periods are identified as wet and dry seasons. The wet season is defined as October 1 to May 30 and the dry season is from May 31 to September 30.

1.4 Description of Sampling Media and Techniques

Four sample types were collected in accordance with guidelines set forth in the Sampling Plan. The four sampling types include: tree wood chips, leaf samples, duff samples, and plant-transpired water samples. A brief description of the procedure for collection of each sample type is included below.

Tree wood chips

Tree wood chips were collected from the specified trees by drilling from the bark to the center of the tree. The chips created by the drill bit were collected in a labeled plastic bag. Full borehole lengths were drilled to ensure that the sample was representative of the tree cross-section. The plastic bag of chips was sealed and placed in an ice chest or refrigerator until shipment to the analytical laboratory.

Leaf samples

Leaf samples were collected from low-hanging branches near where the tree wood chips were collected. Leaves were removed from the tree by hand and placed into a labeled bag. The bag was sealed and stored in an ice chest or refrigerator until shipment to the analytical laboratory.

Duff samples

Duff is material such as leaves, bark, and twigs that accumulates on the ground. Duff samples were collected by reaching into the duff layer (within 5 meters of the tree sampled for wood chips), picking up a sample, and placing the sample into a plastic bag. Like wood chips and leaves, the duff samples were placed on ice or in a refrigerator until they were shipped for analysis.

Plant-transpired water samples

Samples of plant-transpired water were obtained by securing a plastic bag completely over a branch of the tree. An air- and water-tight seal was formed by taping the bag securely to the branch and placing a rubber band over the bag and the stem. All water transpired by the branch was captured in the bag and transferred to a glass bottle for laboratory analysis.

1.5 Analytical Techniques for Measuring TFWT and OBT

The vegetation samples taken in September and November of 2001 were sent to two laboratories for analysis. Samples from each of the ten locations were sent to Eberline Services Laboratory in Richmond, CA. At one location, duplicate samples were collected and sent to Eberline

Services Laboratory. Split samples from a selected set of locations were sent to an EPA contract laboratory, the Center for Applied Isotope Studies (CAIS) at the University of Georgia.

The contracted required minimum detectable activities (MDA) specified in the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001) include 0.50 pCi/g for TFWT in vegetation samples, 200 pCi/g for TFWT in plant-transpired water, and 5.0 pCi/g for OBT in vegetation samples. In all cases, the actual limits achievable by both laboratories analyzing LBNL vegetation samples were lower than the contracted required limits imposed by the Vegetation Sampling Plan for Tritium.

Eberline Services Analytical Technique

The vegetation samples were analyzed for TFWT by Eberline Services using azeotropic distillation. Azeotropic distillation allows low-temperature removal of water without destroying organic material. The vegetation samples were analyzed for OBT by Eberline Services using an oxidation technique. Eberline analyzed the tritiated water samples for TFWT using simple distillation and liquid scintillation techniques.

Center for Applied Isotope Studies (CAIS) Analytical Technique

The Center for Applied Isotope Studies (CAIS) at the University of Georgia removed adsorbed water from the samples under vacuum. For the samples designated for OBT analysis, an aliquot of the sample was dried in a laboratory oven before checking for weight loss. An aliquot of the dry sample was weighed and loaded into a “Parr Bomb” sample cup. A “Parr Bomb” is a sealed container filled to 300 psig with pure dry oxygen. Ignition was effected electrically. After the pressure stabilized in the bomb, the contents were drawn through a trap immersed in dry ice-isopropanol slush and the water from the oxidation was thus condensed. Ultra low-background liquid scintillation counters were used by CAIS for measurement.

2.0 RESULTS

The results of the 2001 sampling are presented in Tables 2 through 6 and Figures 2 and 3. Measurements of TFWT and OBT in leaves, wood chips, duff, and plant-transpired water are presented, along with their associated measurement uncertainty, for two sampling periods. All results are reported on a wet-weight basis (fresh tissue).

Measurements taken beyond a few hundred meters from the hillside stack are at or below the limits of detection. Although concentrations are detected in a small number of samples further than a few hundred meters from the stack, the concentrations are much smaller than for samples taken near the stack and are very near the limits of detection. For example, the reported concentration of OBT at SEE9 (20,800 m from the hillside stack) is 3.1 pCi/g with a measurement error of 2.0 pCi/g. The detection limit for this sample is 3.0 pCi/g. This apparent detection is not statistically significant because it is very close to the detection limit and has a large associated measurement error (66%). This measurement should be considered a non-detect result.

In addition, the exposure assessment (Section 5.0) is based on the largest concentrations measured; therefore, exposure to tritium measured further from the stack would result in markedly lower doses and risks.

2.1 Tissue Free Water Tritium in Vegetation Samples

For leaf samples collected in September 2001 (Table 2), detectible quantities of TFWT were identified in samples from 6 locations (NNW1, NNW2, NNW3, WNW4, NNN5, and EEE6). For leaf samples collected in November 2001 (Table 3), TFWT was detected in samples from seven locations (NNW1, NNW2, NNW3, WNW4, NNN5, EEE6, and WWW8). All but one leaf sample taken at distances further than 400 meters from the NTLF hillside stack did not contain TFWT at levels that were detectable. The lowest measurable concentration of TFWT in leaves occurred approximately 360 meters from the stack (except WWW8).

For wood samples gathered in September 2001, detectible quantities of TFWT were identified in samples collected from 6 locations (NNW1, NNW2, NNW3, WNW4, NNN5, and EEE6). For wood samples collected in November 2001, detectible quantities of TFWT were identified in samples from 5 locations (NNW1, NNW2, WNW4, NNN5, and EEE6). All wood samples taken at distances further than 400 meters from the NTLF hillside stack did not contain TFWT at levels that were detectable. The lowest measurable concentration of TFWT in wood samples occurred approximately 360 meters from the stack.

Duff samples collected in September 2001 contained detectible quantities of TFWT from 6 locations (NNW1, NNW2, NNW3, WNW4, NNN5, and EEE6). Leaf samples collected in November 2001 contained detectible quantities of TFWT from seven locations (NNW1, NNW2, NNW3, WNW4, NNN5, EEE6, and NEE10). All but one duff sample taken at distances further than 400 meters from the NTLF hillside stack did not contain TFWT at levels that were detectable.

2.2 Tissue Free Water Tritium in Plant-Transpired Water Samples

Measurements of TFWT in vegetation samples collected during the dry season of 2001 (dates of collection: September 12-24, 2001) and the 2001 wet season (dates of collection: November 27, 2001-January 17, 2002) are shown in Table 4. As discussed in the Vegetation Sampling Plan for Tritium (LBNL, 2001), plant-transpired water was sampled at three locations in the NNW direction and at two background locations.

Plant-transpired water samples collected both in September 2001 and January 2002 contained detectible quantities of TFWT at 3 locations (NNW1, NNW2, and NNW3). All samples taken at distances further than approximately 363 meters from the NTLF hillside stack did not contain TFWT at levels that were detectable.

Table 2. Measurements of tissue free water tritium (TFWT) in vegetation samples collected during the dry season of 2001 (dates of collection: September 12-13).

Sample ID	**Distance from NTLF stack**	**Concentration (pCi/g)** Leaves	Wood	Duff
NNW1	27	8.9 ± 0.9 (6.0 ± 0.6) [a]	9.6 ± 1.0 (8.4 ± 0.9)	1.2 ± 0.1 (1.7 ± 0.2)
NNW2	105 ± 12	2.6 ± 0.3 [5.1 ± 0.11] [b]	2.2 ± 0.2 [1.7 ± 0.07]	1.0 ± 0.1 [2.1 ± 0.04]
NNW3	363 ± 7	*0.26 ± 0.08* [c]	*0.16 ± 0.07*	*0.45 ± 0.08*
WNW4	103 ± 10	2.5 ± 0.3	0.80 ± 0.12	1.5 ± 0.2
NNN5	45 ± 6	5.1 ± 0.5	4.2 ± 0.4	1.4 ± 0.2
EEE6	215 ± 8	0.73 ± 0.11	*0.19 ± 0.07*	*0.14 ± 0.05*
SSE7	529 ± 8	*<0.14* [d]	*<0.14*	*<0.11*
WWW8	832 ± 34	*<0.14*	*<0.14*	*<0.10*
SEE9	20,800 ± 12	*<0.14*	*<0.14*	*<0.10*
NEE10	1080 ± 12	*<0.14*	*<0.14*	*<0.10*

[a] Values in parentheses ( ) represent results of duplicate sample analyses performed by Eberline Services, Richmond, CA.

[b] Values in brackets [ ] represent results of split sample analyses performed by the Center for Applied Isotope Studies (CAIS) at the University of Georgia.

[c] Values in *italics* represent measurements that were less than the contracted required minimum detectable activity (MDA) specified in the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001). The contracted required limit for TFWT is 0.5 pCi/g.

[d] Less than (<) values indicate that the concentration of tritium in the sample was below the analytical detection limit achievable by the laboratory.

Table 3. Measurements of tissue free water tritium (TFWT) in vegetation samples collected during the wet season of 2001 (dates of collection: November 27-29).

Sample ID	Distance from NTLF stack	Concentration (pCi/g) Leaves	Wood	Duff
NNW1	27	9.0 ± 0.9 (9.2 ±1.0) [a] [21 ± 0.3] [b]	8.7 ± 0.9 (9.5 ± 1.0) [10 ± 0.2]	9.3 ± 1.0 (9.3 ±1.0) [6.6 ± 0.2]
NNW2	105 ± 12	4.1 ± 0.4 [2.4 ± 0.06]	1.8 ± 0.2 [2.0 ± 0.09]	3.6 ± 0.4 [2.6 ± 0.1]
NNW3	363 ± 7	*0.41 ± 0.10* [c]	*<0.13*	*0.12 ± 0.06*
WNW4	103 ± 10	1.3 ± 0.2	0.62 ± 0.12	1.4 ± 0.2
NNN5	45 ± 6	5.0 ± 0.5 [19 ± 0.2]	4.5 ± 0.5 [4.5 ± 0.1]	4.6 ± 0.5 [3.7 ± 0.1]
EEE6	215 ± 8	*0.36 ± 0.1*	*0.21* ± 0.08	0.59 ± 0.12
SSE7	529 ± 8	*<0.13* [d]	*<0.14*	*<0.13*
WWW8	832 ± 34	*0.24 ± 0.1*	*<0.12*	*<0.14*
SEE9	20,800 ± 12	*<0.12*	*<0.12*	*<0.10*
NEE10	1080 ± 12	*<0.13*	*<0.13*	*0.22 ± 0.1*

[a] Values in parentheses ( ) represent results of duplicate sample analyses performed by Eberline Services, Richmond, CA.

[b] Values in brackets [ ] represent results of split sample analyses performed by the Center for Applied Isotope Studies (CAIS) at the University of Georgia.

[c] Values in *italics* represent measurements that were less than the contracted required minimum detectable activity (MDA) specified in the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001). The contracted required limit for TFWT is 0.5 pCi/g.

[d] Less than (<) values indicate that the concentration of tritium in the sample was below the analytical detection limit achievable by the laboratory.

Table 4. Measurements of tissue free water tritium (TFWT) in plant-transpired water collected in 2001 (dry season collection dates: September 19-24, 2001; wet season collection dates: January 3-17, 2002).

Sample ID [a]	**Distance from NTLF stack**	**Concentration (pCi/mL)** Dry season	Wet season
NNW1	27	13.6 ± 1.4 (13.0 ± 1.3) [b]	8.8 ± 0.9 (11.4 ± 1.2) [9.6 ± 0.3] [c]
NNW2	105 ± 12	3.7 ± 0.4 [3.7 ± 0.1]	3.9 ± 0.4 [3.8 ± 0.2]
NNW3	363 ± 7	0.43 ± 0.13	0.38 ± 0.13
SEE9	20,800 ± 12	*<0.18* [d,e]	*<0.18*
NEE10	1080 ± 12	*<0.18*	*<0.18*

[a] In accordance with the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001), sampling of plant-transpired water was limited to five locations.

[b] Values in parentheses ( ) represent results of duplicate sample analyses performed by Eberline Services, Richmond, CA.

[c] Values in brackets [ ] represent results of split sample analyses performed by the Center for Applied Isotope Studies (CAIS) at the University of Georgia.

[d] Values in *italics* represent measurements that were less than the contracted required minimum detectable activity (MDA) specified in the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001). The contracted required limit for TFWT in plant-transpired water is 0.20 pCi/L.

[e] Less than (<) values indicate that the concentration of tritium in the sample was below the analytical detection limit achievable by the laboratory.

Table 5. Measurements of organically bound tritium (OBT) in vegetation samples collected during the dry season of 2001 (dates of collection: September 12-13).

	Distance from	**Concentration (pCi/g)**		
Sample ID	**NTLF stack**	Leaves	Wood	Duff
NNW1	27	41 ± 5 (49 ± 6) [a]	*4.3 ± 2.6* [b] (*<4.0*)	240 ± 25 (290 ± 30)
NNW2	105 ± 12	24 ± 4 [16 ± 0.1] [c]	*1.7 ± 2.3* [*0.81 ± 0.04*]	24 ± 5 [38 ± 0.2]
NNW3	363 ± 7	*<3.0* [d]	*<4.0*	*<4.0*
WNW4	103 ± 10	18 ± 3	*<3.0*	13 ± 3
NNN5	45 ± 6	30 ± 4	*<3.0*	84 ± 10
EEE6	215 ± 8	7.1 ± 2.1	*4.7 ± 2.4*	15 ± 4
SSE7	529 ± 8	*4.6 ± 1.9*	*<3.0*	*<4.0*
WWW8	832 ± 34	*<3.0*	*<3.0*	*<4.0*
SEE9	20,800 ± 12	*3.1 ± 2.0*	*<3.0*	*<4.0*
NEE10	1080 ± 12	*<3.0*	*<4.0*	*<4.0*

[a] Values in parentheses ( ) represent results of duplicate sample analyses performed by Eberline Services, Richmond, CA.

[b] Values in *italics* represent measurements that were less than the contracted required minimum detectable activity (MDA) specified in the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001). The contracted required limit for OBT is 5.0 pCi/g.

[c] Values in brackets [ ] represent results of split sample analyses performed by the Center for Applied Isotope Studies (CAIS) at the University of Georgia.

[d] Less than (<) values indicate that the concentration of tritium in the sample was below the analytical detection limit achievable by the laboratory.

Table 6. Measurements of organically bound tritium (OBT) in vegetation samples collected during the wet season of 2001 (dates of collection: November 27-29).

	Distance from	**Concentration (pCi/g)**		
Sample ID	**NTLF stack**	Leaves	Wood	Duff
NNW1	27	29 ± 4 (45 ± 6) [a] [39 ± 0.2] [b]	5.4 ± 3 (*<4.0*) [c] [3.9 ± 0.1]	120 ± 13 (140 ± 15) [130 ± 0.4]
NNW2	105 ± 12	23 ± 4 [19 ± 0.2]	*<4.0* [*1.0 ± 0.04*]	24 ± 4 [57 ± 0.5]
NNW3	363 ± 7	*<4.0* [d]	*<4.0*	*<4.0*
WNW4	103 ± 10	25 ± 4	*<4.0*	12 ± 4
NNN5	45 ± 6	40 ±6 [16 ± 0.1]	*<4.0* [*1.1 ± 0.05*]	78 ± 9 [62 ± 0.3]
EEE6	215 ± 8	10 ± 3	<5.0	9.4 ± 3.2
SSE7	529 ± 8	*<4.0*	*<4.0*	*<4.0*
WWW8	832 ± 34	*<4.0*	*<4.0*	*<4.0*
SEE9	20,800 ± 12	*<4.0*	*<4.0*	*<4.0*
NEE10	1080 ± 12	*<4.0*	*<4.0*	*<5.0*

[a] Values in parentheses ( ) represent results of duplicate sample analyses performed by Eberline Services, Richmond, CA.

[b] Values in brackets [ ] represent results of split sample analyses performed by the Center for Applied Isotope Studies (CAIS) at the University of Georgia.

[c] Values in *italics* represent measurements that were less than the contracted required minimum detectable activity (MDA) specified in the 2001 Vegetation Sampling Plan for Tritium (LBNL, 2001). The contracted required limit for OBT is 5.0 pCi/g.

[d] Less than (<) values indicate that the concentration of tritium in the sample was below the analytical detection limit achievable by the laboratory.

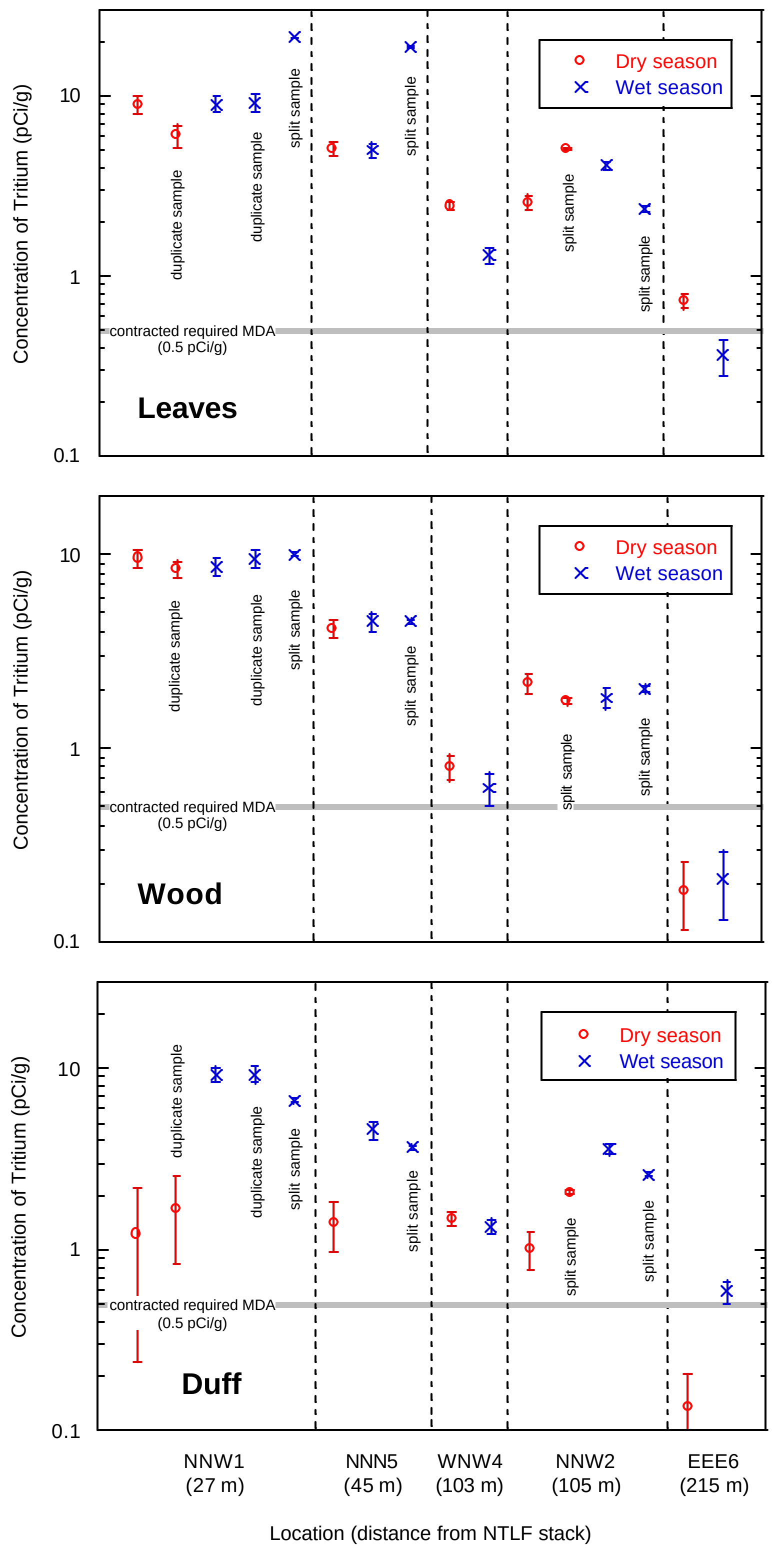

Figure 2. Concentration of TFWT in vegetation near the NTLF stack; samples collected in September 2001 (dry season) and November 2001 (wet season).

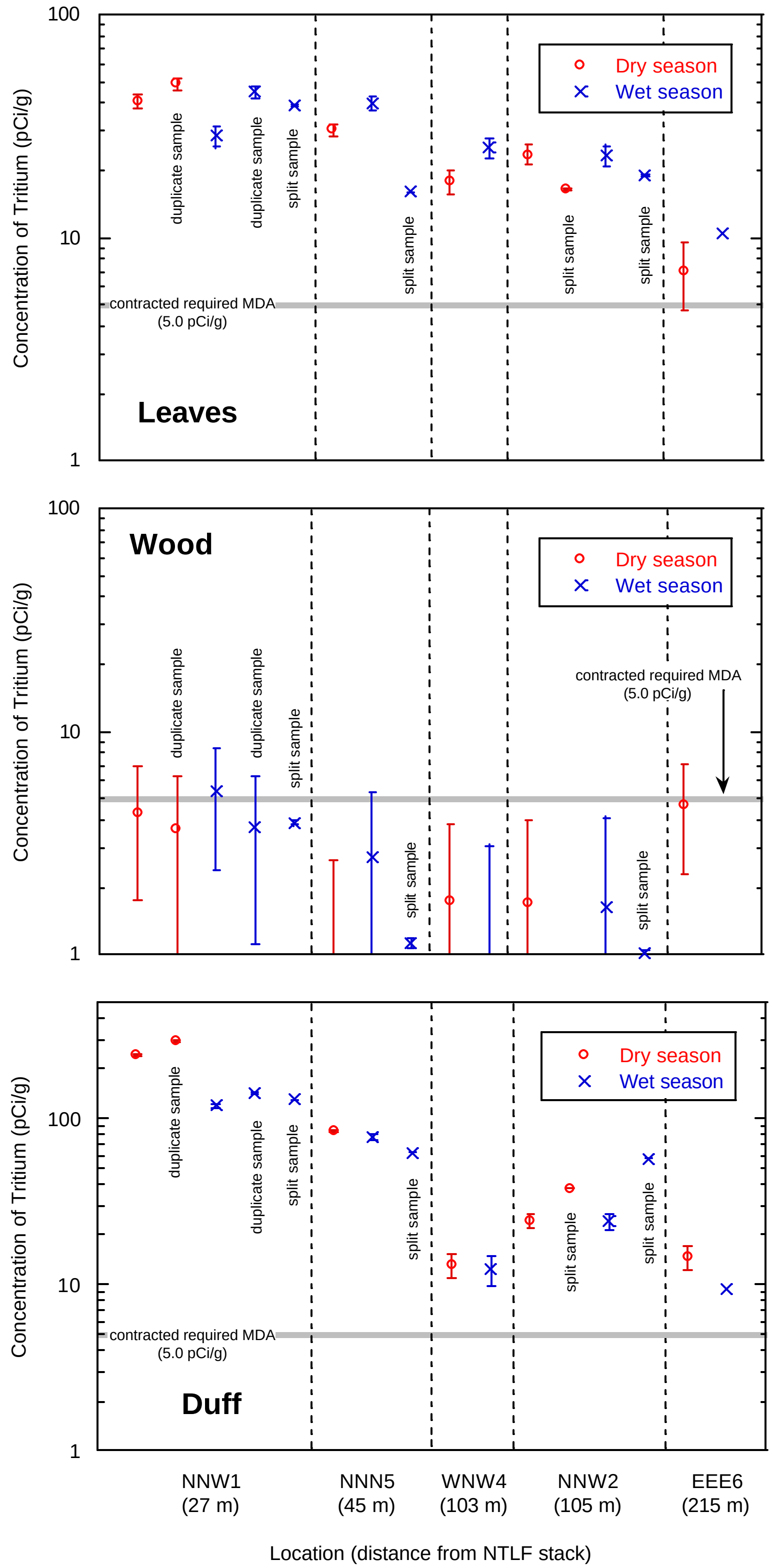

Figure 3. Concentration of OBT in vegetation near the NTLF stack; samples collected in September 2001 (dry season) and November 2001 (wet season).

2.3 Organically Bound Tritium in Vegetation Samples

Measurements of OBT in vegetation samples collected during the dry season of 2001 (dates of collection: September 12-13) are shown in Table 5; samples collected during the 2001 wet season (dates of collection: November 27-29) are presented in Table 6.

For leaf and duff samples collected both in September 2001 and November 2001, detectible quantities of OBT were identified in samples from 5 locations (NNW1, NNW2, WNW4, NNN5, and EEE6). All measurements of OBT in tree wood, except one (NNW1; November 27-29, 2001), were lower than the contracted required minimum detectable activity (MDA) of 5.0 pCi/g specified in the 2001 Vegetation Sampling Plan for Tritium.

2.4 Discussion of 2001 Results

The results of the 2001 vegetation sampling indicate that differences in season (wet vs. dry) do not cause a major fluctuation in the concentration of tritium in vegetation. The only situation that appears to be affected by the time of the year that sampling occurs is TFWT in duff. The increased concentration of TFWT in duff during the wet season could be indicative of rain scavenging HTO vapor in the air column above ground leading to enhanced deposition of HTO on materials that comprise duff at ground level. The fact that OBT in duff does not change from season to season suggests that the differences in TFWT in duff between the dry and wet season could be due to rainfall scavenging of HTO in air and in foliage above ground level.

The concentration of OBT in leaves and duff (Tables 5 and 6) is larger than the concentration of TFWT in leaves and duff (Tables 2 and 3). This is an indication that the OBT measured in the leaves and duff is a result of tritium released over the past several years as opposed to tritium currently being released.

Tables 7 through 9 show that most samples taken in 1998 indicated higher concentrations of OBT and TFWT than corresponding samples taken in 2001. This downwards trend in concentration of tritium in vegetation is likely a result of reduced amounts of tritium released from the NTLF since 1998 (Figure 4).

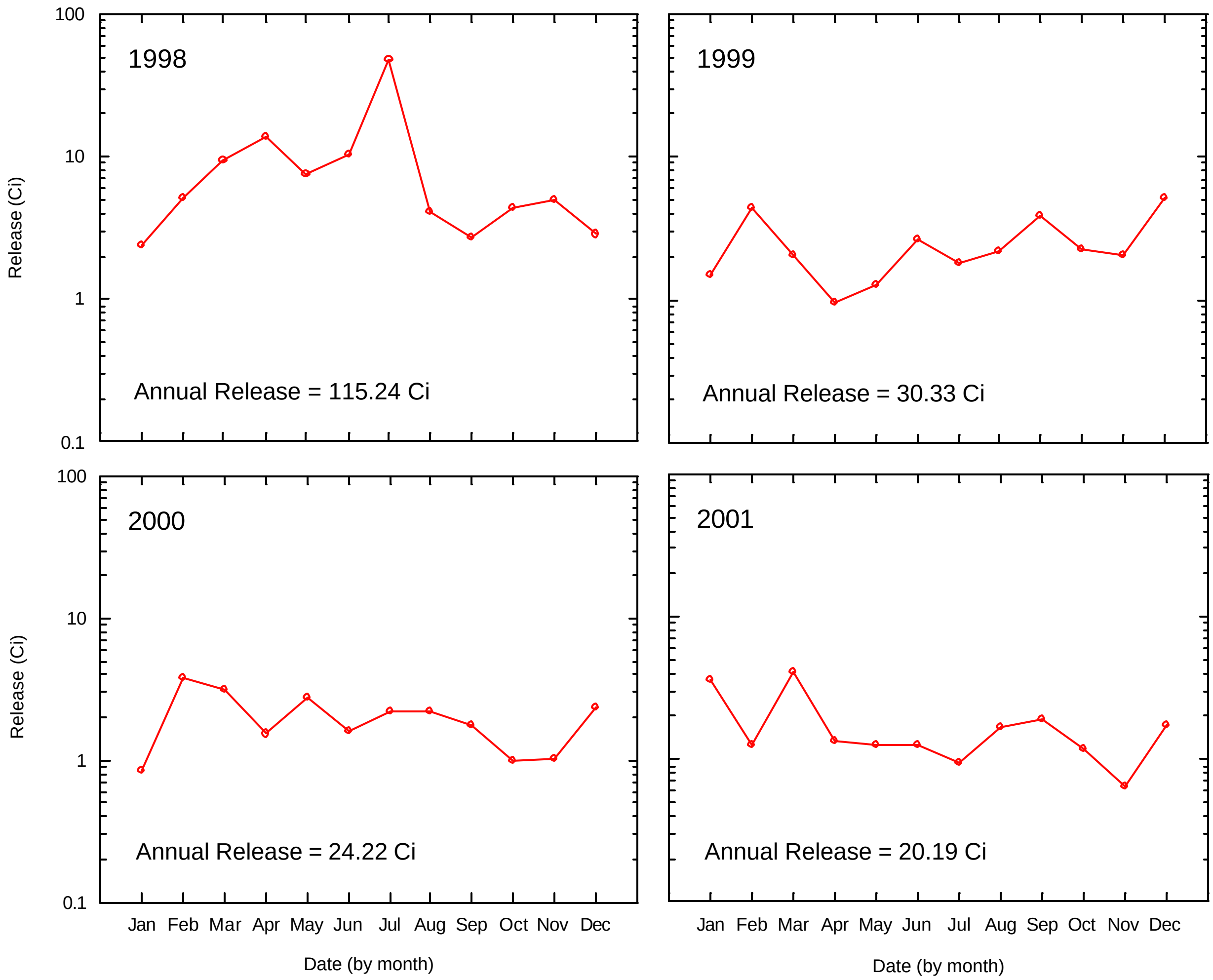

Figure 4. Measured releases of tritium from the National Tritium Labeling Facility from 1998 through 2001.

3.0 COMPARISON OF 2001 RESULTS WITH PAST MEASUREMENTS

This section contains a comparison of samples taken in September and November of 2001 with samples taken in previous years. Tables 7, 8, and 9 present comparisons for leaves, wood chips, and duff, respectively. All samples are given on a wet-weight basis. Although many vegetation samples have been collected and analyzed over the past several years, this report discusses and compares only the samples collected at distances and directions similar to the ten locations discussed in Section 1.1.

Measurements of TFWT in leaves taken in 1998 do not vary significantly from measurements taken at comparable locations in 2001. However, estimates of OBT in leaves were higher for all compared locations in 1998 than in 2001, possibly indicating higher exposures to TFWT in the past which led to higher rates of transformation from TFWT to OBT.

TFWT concentrations in wood chip samples collected in 1998 were generally larger (approximately a factor of 2) than wood chip samples collected in 2001. Significant conclusions cannot be drawn regarding OBT in wood chips because the OBT content in wood chips is at or below the limits of detection beyond 40 m of the NTLF stack.

A comparison of TFWT in duff samples shows that measurements taken in 2001 are essentially equivalent to 1998 measurements. However, seasonal effects are seen in the 2001 TFWT results. The concentrations of OBT in 1998 duff samples were higher in all cases than in the 2001 samples. This is likely indicative of higher releases occurring in the past.

Table 7. Comparison of Leaves Samples taken in 2001 with Samples taken in Previous Years.

Analysis type	Direction from NTLF stack	Distance from NTLF stack (m)	Date of collection	Concentration (pCi/g)
TFWT	NNW	20	9/16/98	6.3 ± 0.7
	NNW	27	9/13/01	8.9 ± 0.9
	NNW	27	11/29/01	9.0 ± 0.9
	NNW	46	9/17/98	8.0 ± 0.8
	N	45	9/13/01	5.1 ± 0.5
	N	45	11/29/01	5.0 ± 0.5
	NNW	97	9/17/98	4.4 ± 0.5
	NNW	105	9/13/01	2.6 ± 0.3
	NNW	105	11/29/01	4.1 ± 0.4
	SE	20,000	9/18/98	<0.10
	SE	20,800	9/12/01	<0.14
	SE	20,800	11/27/01	<0.12
OBT	NNW	20	9/16/98	68 ± 7
	NNW	27	9/13/01	41 ± 5
	NNW	27	11/29/01	29 ± 4
	NNW	46	9/17/98	72 ± 8
	N	45	9/13/01	30 ± 4
	N	45	11/29/01	40 ± 6
	NNW	97	9/17/98	50 ± 5
	NNW	105	9/13/01	24 ± 4
	NNW	105	11/29/01	23 ± 4
	SE	20,000	9/18/98	<1.5
	SE	20,800	9/12/01	<3.1
	SE	20,800	11/27/01	<3.9

Table 8. Comparison of Wood Chip Samples taken in 2001 with Samples taken in Previous Years.

Analysis type	Direction from NTLF stack	Distance from NTLF stack (m)	Date of collection	Concentration (pCi/g)
TFWT	NNW	25	9/16/98	17 ± 2
	NNW	27	9/13/01	9.6 ± 1.0
	NNW	27	11/29/01	8.7 ± 0.9
	WNW	94	9/15/98	2.1 ± 0.09
	WSW	103	9/13/01	0.80 ± 0.12
	WSW	103	11/29/01	0.62 ± 0.12
	NNW	97	9/17/98	2.3 ± 0.3
	NNW	105	9/13/01	2.2 ± 0.2
	NNW	105	11/29/01	1.8 ± 0.2
	SE	20,000	9/18/98	<0.10
	SE	20,800	9/12/01	<0.14
	SE	20,800	11/27/01	<0.12
OBT	NNW	20	9/16/98	3.6 ± 1.2
	NNW	27	9/13/01	4.3 ± 2.6
	NNW	27	11/29/01	5.4 ± 3
	NNW	46	9/15/98	<1.4
	N	45	9/13/01	<3.1
	N	45	11/29/01	<4.2
	NNW	97	9/16/98	<1.8
	NNW	105	9/13/01	1.7 ± 2.3
	NNW	105	11/29/01	<4.1
	SE	20,000	9/18/98	<1.8
	SE	20,800	9/12/01	<3.2
	SE	20,800	11/27/01	<4.2

Table 9 Comparison of Duff Samples taken in 2001 with Samples taken in Previous Years.

Analysis type	Direction from NTLF stack	Distance from NTLF stack (m)	Date of collection	Concentration (pCi/g)
TFWT	NNW	20	9/16/98	6.4 ± 0.7
	NNW	27	9/13/01	1.2 ± 0.1
	NNW	27	11/29/01	9.3 ± 1.0
	N	50	3/30/99	7.7 ± 0.8
	N	45	9/13/01	1.4 ± 0.2
	N	45	11/29/01	4.6 ± 0.5
	NNW	97	9/17/98	3.0 ± 0.3
	NNW	105	9/13/01	1.0 ± 0.1
	NNW	105	11/29/01	3.6 ± 0.4
	SE	20,000	9/18/98	0.23 + 0.06
	SE	20,800	9/12/01	<0.10
	SE	20,800	11/27/01	<0.10
OBT	NNW	20	9/16/98	1300 ± 130
	NNW	20	3/31/99	180 + 19 [a]
	NNW	27	9/13/01	240 ± 25
	NNW	27	11/29/01	120 ± 13
	NNW	46	9/15/98	250 ± 25
	N	45	9/13/01	84 ± 10
	N	45	11/29/01	78 ± 9
	NNW	97	9/17/98	76 ± 8
	NNW	105	9/13/01	24 ± 5
	NNW	105	11/29/01	24 ± 4
	SE	20,000	9/18/98	<2.7
	SE	20,800	9/12/01	<4.3
	SE	20,800	11/27/01	<3.8

[a] Average of 4 samples taken at this location on 3/31/99.

4.0 COMPARISON WITH CONCENTRATIONS ESTIMATED IN 1997 BY MCKONE

An environmental health-risk assessment for tritium (McKone, 1997) estimated risk to the population from tritium released from LBNL. The risk assessment considered three regions (zones) in which people might be exposed to tritium from LBNL. The borders of zones 1, 2, and 3 are concentric circles with radii of 100, 1100, and 2100 meters, respectively.

The McKone study estimated the average concentration of free water tritium in vegetation in zone 2 (the region that encompasses most sampling locations discussed in this report) to be 110 Bq/kg fresh mass (3 pCi/g). The predicted concentration of OBT in vegetation for the same zone is 3 Bq/kg (0.08 pCi/g). These estimates assume an annual release of 100 Ci.

A direct comparison between the McKone estimates and the measurements presented in this report is difficult due to the following reasons: 1) the McKone estimates assume an annual release of 100 Ci and are averaged over a large region (out to 1100 meters from the NTLF hillside stack) while the measurements are taken at specific locations under conditions when annual emissions decreased from about 30 to 20 Ci/yr from 1999 to 2001 (Figure 4) and 2) the McKone predictions are for vegetation in general while the measurements are for specific vegetation types (leaves, tree wood, and duff).

Nevertheless, McKone's predictions appear reasonable compared to the measurements in this report. McKone's predictions for zone 2 (100 m to 1100 m from the hillside stack) are similar to measurements taken in this region. The measurements of TFWT are lower than McKone's predictions. This is reflective of McKone's assumption that 100 Ci are released per year. The release rate at the time of the 2001 measurements was approximately 20 Ci/yr (Figure 4). Since the concentration of OBT in vegetation is at or below limits of detection, it is not possible to compare with the McKone prediction for OBT, other than to say that they are both consistently small.

Table 10. Comparison of predictions of tritium concentrations in vegetation (McKone, 1997) and measured concentrations in wood taken in 1998 and 2001.

Analysis type	Direction from NTLF stack	Distance from NTLF stack (m)	Date	Concentration (pCi/g)
TFWT	WNW	94	9/15/98	2.1 ± 0.09
	WSW	103	9/13/01	0.80 ± 0.12
	WSW	103	11/29/01	0.62 ± 0.12
	NNW	97	9/17/98	2.3 ± 0.3
	NNW	105	9/13/01	2.2 ± 0.2
	NNW	105	11/29/01	1.8 ± 0.2
McKone prediction		100 to 1000	1997	3.0
OBT	NNW	97	9/16/98	<1.8
	NNW	105	9/13/01	1.7 ± 2.3
	NNW	105	11/29/01	<4.1
McKone prediction		100 to 1000	1997	0.08

5.0 ASSESSMENT OF POTENTIAL RADIATION DOSES AND HEALTH RISKS

Tritium in tree wood and other vegetation can be released to the atmosphere, surface water, or soil. Releases of tritium to the environment can lead to exposures of humans. This section considers potential radiation doses and associated health risks to the public due to the presence of tritium in tree wood, leaves, and groundcover (duff) in the vicinity of the NTLF.

The assessment of potential doses and health risks to the public is based in part on an analysis currently being performed as part of an application for an authorized release limit for tritium in tree wood and other vegetative materials (LBNL, draft report). A recent assessment of radiation doses to the public from disposal of tritium in consumer products and other items (Schneider et al., 2001) also is used.

5.1 Assessment of Potential Doses

As part of the analysis in the draft LBNL report, hypothetical scenarios for exposure to tritium in tree wood and other vegetative materials were identified, and individual and collective (population) doses in these scenarios were estimated. The assumed exposure scenarios include: (1) exposure of workers during processing of wood into paper products, (2) exposure of workers during processing of wood and leaves into mulch, (3) exposure during use of wood in domestic fireplaces, and (4) exposure of the public following disposal of wood, leaves, and duff in a landfill. Other exposure scenarios could be considered. However, the scenarios that have been evaluated should provide reasonable representations of exposure situations for which the potential doses and health risks are the highest.

Results of dose analyses for the different exposure scenarios considered in the draft LBNL report are summarized in Table 11. Results for the first three scenarios are obtained from the draft report. However, results for the scenario involving disposal of vegetation in a landfill are obtained from an analysis by Schneider et al. (2001). Estimates of individual and collective dose are normalized to an activity concentration of tritium in vegetation of 1 picocurie (pCi) per gram. The dose estimates for disposal of vegetation in a landfill are based on assumptions that no more than 10% of the total vegetation mass in the vicinity of the NTLF would be removed in any year and that the release rate of tritium from vegetation after disposal would be 10% of the release rate from loose materials. The estimated individual dose is the highest in the scenario involving processing of vegetation into mulch, whereas the estimated collective dose is the highest in the scenario involving use of wood in domestic fireplaces, due to the much larger population of firewood users compared with assumed populations of workers at a paper mill or mulching facility.

The estimates of individual and collective dose per unit concentration of tritium in vegetation in the first three scenarios in Table 11 probably are very conservative. These estimates are based, in part, on an assumption that either 10% or 50% of the water vapor in air at 50% or 100% relative humidity is tritiated water released during processing of the vegetation. This assumption should result in large overestimates of concentrations of tritium in air and, thus, doses from inhalation and absorption through the skin. In the scenario involving disposal of vegetation in a landfill, the assumed release rate of tritium from wood compared with the release rate from loose materials also should be conservative, given the slow rate of decomposition of wood after disposal.

Table 11. Estimates of individual and collective (population) doses per unit activity concentration of tritium in wood and vegetative material

Assumed disposition of wood and vegetative material	Material	Individual dose (mrem per pCi/g)	Collective dose (person-rem per pCi/g)
Processing of wood into paper products[a]	Wood	3.8×10^{-5}	1.9×10^{-5}
Processing of wood and leaves into mulch[a]	Wood, leaves	2.9×10^{-4}	2.9×10^{-6}
Use of wood in domestic fireplaces[a]	Wood	1.4×10^{-5}	1.2×10^{-4}
Disposal in landfill[b]	Wood	1.3×10^{-7}	3.0×10^{-8}

[a] Estimates based on analysis given in LBNL (draft report).
[b] Estimates based on analysis given in Appendix A.2 of Schneider et al. (2001).

Potential individual and collective doses to the public can be estimated from the data on concentrations of tritium in vegetation summarized in Tables 2, 3, 5, and 6 and the estimates of individual and collective dose per unit concentration in Table 11. In samples taken during 2001, the maximum concentration of tritium in wood chips is about 15 pCi/g and the maximum concentration of tritium in leaves is about 290 pCi/g. These values are the sum of concentrations of tissue free water tritium (TFWT) and organically bound tritium (OBT). A distinction between the two kinds of tritium is not needed because the exposure scenarios assume that all tritium released to the air is in the form of tritiated water vapor. The following estimates of dose are obtained.

- The maximum individual dose, which occurs in the scenario involving processing of wood and leaves into mulch, is about 8×10^{-2} mrem.
- The maximum collective dose, which occurs in the scenario involving use of wood in domestic fireplaces, is about 2×10^{-3} person-rem.

Again, these results should be very conservative estimates of doses that would be received as a result of the assumed uses of vegetation contaminated with tritium.

The estimated individual and collective doses summarized above are very low. The maximum individual dose of 8×10^{-2} mrem is only 0.8% of the current dose limit for airborne releases of radionuclides of 10 mrem per year specified by the U.S. Environmental Protection Agency in 40 CFR Part 61. The maximum collective dose of 2×10^{-3} person-rem is only 0.02% of the guideline used by the U.S. Department of Energy (DOE, 1991) to define collective doses that are as low as reasonably achievable (ALARA). Both of these doses are far below levels of concern for any exposure situation involving radionuclides in the environment.

5.2 Assessment of Potential Health Risks

Estimates of health risks associated with the doses given above can be obtained based on an assumption that the risk of cancer incidence per unit dose is 1×10^{-3} per rem (Thomas and Hoffman, 2000; Thomas et al., 2000). For a worker at a mulch processing facility, the estimated lifetime risk of cancer incidence would be $(8 \times 10^{-2}$ mrem$)(1 \times 10^{-6}$ per mrem$) = 8 \times 10^{-8}$.

Lifetime cancer risks to individuals in the other exposure scenarios would be lower. The estimated risk, although it is likely to be a large overestimate of the actual risk to a worker, is far below levels of concern for any exposure situation involving cancer-causing substances in the environment. Such risks are far too small to be observable, even if a very large population were exposed at this level.

For the maximum collective dose in any scenario of about 2×10^{-3} person-rem, which is the estimate for use of wood in domestic fireplaces, using an assumed risk per unit dose of 1×10^{-3} per rem, the expected number of cancers in the exposed population is zero.

5.3 Exposure and Risk from Tritium Released through Natural Transpiration

Exposures and risks also have been estimated for an employee of the LHS exposed to tritium that is released through natural transpiration from the hillside grove of eucalyptus trees. Conservative bounding calculations have been performed as a surrogate to the use of an area source dispersion model.

The first step in this calculation is to estimate the amount of tritium that is being released to the environment from the tree grove. The rate of transpiration in eucalyptus trees is approximately 40 liters per day per tree (FAO, 1998). Assuming there are 300 trees in the grove gives 40 L/d/tree × 300 trees = 12,000 liters of water per day being transpired by the entire grove. Multiplying this transpiration rate times the largest measured concentration of plant-transpired water of 13.6 pCi/mL (see Table 4) gives a release of 0.06 Ci/y from the grove. This release is a small fraction of the release of 20 to 30 Ci/y from the NTLF hillside stack from 1999 through 2001.

Tritium released from the trees is mixed with air that flows through the grove. A dispersion factor is used to estimate the amount of air during the course of a year that would mix with the transpired HTO in leaves to produce an average concentration in air at the edge of the grove. The dispersion factor is estimated as the inverse of the product of the area of the grove (100 m × 200 m = 20,000 m^2), the height of the grove (50 m), and the turnover rate per second of air in the grove. The air turnover rate is estimated from an average wind speed and an assumption that the wind direction is along the length of the grove (200 m). For an assumed average wind speed of

2 m/s, the air turnover rate is (2 m/s)/(200 m) = $0.01\ s^{-1}$. The dispersion factor then is 1/(20,000 $m^2 \times 50\ m \times 0.01\ s^{-1} = 1 \times 10^{-4}\ s/m^3$.

The concentration of tritium in air resulting from transpiration from the grove is estimated by multiplying the release rate (0.06 Ci/y) by the dispersion factor ($1 \times 10^{-4}\ s/m^3$). After accounting for conversion of units, the air concentration is 0.2 pCi/m^3.

The lifetime dose and risk has been estimated (Table 12) for an employee of the LHS working for 30 years, beginning at age 20, using the methodology and assumptions discussed in a radiological assessment completed in 2000 for LHS workers (Thomas et al., 2000).

The dose was estimated for an LHS employee potentially exposed to tritium originating from the hillside grove for 9 hours per day, 5 days per week, for 52 weeks (2,340 hours/year), minus vacation, holidays and sick-leave (approximately 33 days/year). Therefore, it is assumed that the employee spends 2043 work-day hours per year at the LHS.

The indoor air tritium concentration at the LHS is assumed to be equivalent to those calculated for the southern patio of the LHS. The intake of HTO into the body is assumed to occur primarily from inhalation and skin absorption. The breathing rate (1.4 m^3/h) used in this analysis is based on studies presented in EPA's Exposure Factors Handbook (EPA, 1997).

The estimated dose that an employee of the LHS for 30 years would receive from exposure to tritium being released from the hillside grove through transpiration would be less than 0.004 mrem. The lifetime risk from this dose would be less than 4×10^{-9}; calculated as (0.004 mrem)(1×10^{-6} risk/mrem). Because of the conservative assumptions used to produce these results, they should most appropriately be interpreted as dose and risk estimates that would not likely be exceeded by exposures to real persons. For this reason, the above values are referred to as "less than" values.

The conservative assumptions used in this analysis include: (a) the largest measured concentration of HTO in transpired water was assumed to be the average, (b) the assumption that the NTLF will continue operations for 30 years, and (c) there is no dilution between the edge of the grove and the LHS.

6.0 CONCLUSIONS

Measurements taken in accordance with the Sampling Plan for Tritium were used in an exposure assessment that considered plausible scenarios that could result in human exposure. This assessment shows that levels of tritium in vegetation near the Berkeley Lab do not pose a health hazard to the public.

No reasonable situations can be conceived of whereby the reported concentrations of tritium in vegetation surrounding LBNL would indicate exposures and/or doses approaching or exceeding a few mrem over a human lifetime. The maximum individual dose, which occurs in the scenario involving processing of wood and leaves into mulch, is about 8×10^{-2} mrem. The maximum collective dose, which occurs in the scenario involving use of wood in domestic fireplaces, is about 2×10^{-3} person-rem. Based on these doses, the expected number of cancers from exposure to tritium in the exposed population is zero.

Conservative bounding calculations were completed to estimate the amount of tritium being released from the hillside grove through transpiration and the resulting dose and risk for a maximally exposed individual at the LHS. The estimated dose that an employee of the LHS for 30 years would receive from exposure to tritium being released from the hillside grove through transpiration would be less than 0.004 mrem. The lifetime risk from this dose would be less than 4×10^{-9}.

In a press release issued by the Environmental Protection Agency (EPA) on July 18, 2002, the EPA announced that environmental sampling at LBNL found tritium levels well below federal health standards and is opting for no further action under the Superfund program (EPA, 2002).

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